REPORT DOCUMENTATION PAGE

AFRL-SR-AR-TR-05-

0022

1. AGENCY USE ONLY *(Leave blank)*	2. REPORT DATE	3. REPOR.
		15 Oct 2003 - 30 Jun 2004 FINAL

4. TITLE AND SUBTITLE
NOVEL TECHNIQUES FOR THE PRODUCTION OF GAMMA RAYS

5. FUNDING NUMBERS
2301/DX
61102F

6. AUTHOR(S)
DR ROBERTS

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
SRS TECHNOLOGIES
1800 QUAIL ST #101
NEWPORT BEACH CA 92660-2301

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES)
AFOSR/NE
4015 WILSON BLVD
SUITE 713
ARLINGTON VA 22203

10. SPONSORING/MONITORING AGENCY REPORT NUMBER
Fa9550-04-C-0006

11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION AVAILABILITY STATEMENT
DISTRIBUTION STATEMENT A: Unlimited

12b. DISTRIBUTION CODE

13. ABSTRACT *(Maximum 200 words)*

Important fundamental research over the past two decades has proven that the energy stored in metastable energy states of some nuclear isomers can be caused to be released from the isomer state by illuminating the nucleus with x-rays. Research has proven that the long-lived naturally abundant isomer of t80Ta can be triggered to release the stored energy by irradiation with Bremsstrahlung x-irradiation. An understanding of this phenomenon within the standard model has now emerged, placing the physics of triggered energy release from isomer states upon a strong experimental and theoretical basis.

The data from the March 2001 Bum-up experiment has now been fully analyzed. No bum-up duc to xynchrotron radiation was detected in this experiment due to both experimental limitations and the unlikelihood of ever observing bum-up in the Hf-i 78m2 system for triggering enhancements of less than 10%. We suggest looking at altemative isomer systems with shorter half-lives, such as the Lu-I 77m isomer.

20050218 078

14. SUBJECT TERMS			15. NUMBER OF PAGES
			16. PRICE CODE
17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
Unclassified	Unclassified	Unclassified	UL

1.0 Background

Important fundamental research over the past two decades has proven that the energy stored in metastable energy states of some nuclear isomers can be caused to be released from the isomer state by illuminating the nucleus with x-rays. Research has proven that the long-lived naturally abundant isomer of ^{180}Ta can be triggered to release the stored energy by irradiation with Bremsstrahlung x-irradiation. An understanding of this phenomenon within the standard model has now emerged, placing the physics of triggered energy release from isomer states upon a strong experimental and theoretical basis.

Investigations of x-ray triggered gamma emissions bear a striking similarity to decades-old research into the excitation of nuclear states, including isomers, by absorption of photons. Both occur through the two-step process schematically depicted in **Figure 1**. A nucleus in a specific initial isomeric state can be excited to reach an intermediate state by absorption of an incident photon. The intermediate state is necessitated by angular momentum considerations – direct electromagnetic transitions between initial and final states are unlikely; otherwise the upper of the two states would not be isomeric. The mediating level must therefore possess values of J (and perhaps K) intermediate between those of initial and final states. In the second step, the intermediate state decays with some branch leading, usually by a cascade, to the final state. The measurable parameters of the reaction are the excitation energy required to reach intermediate state from the initial level and the integral cross section for the overall initial-to-intermediate-to-final-state transfer. Measured values are deduced from the yield of final-state nuclei produced by exposure to driving x-rays, as a function of the energy of those photons.

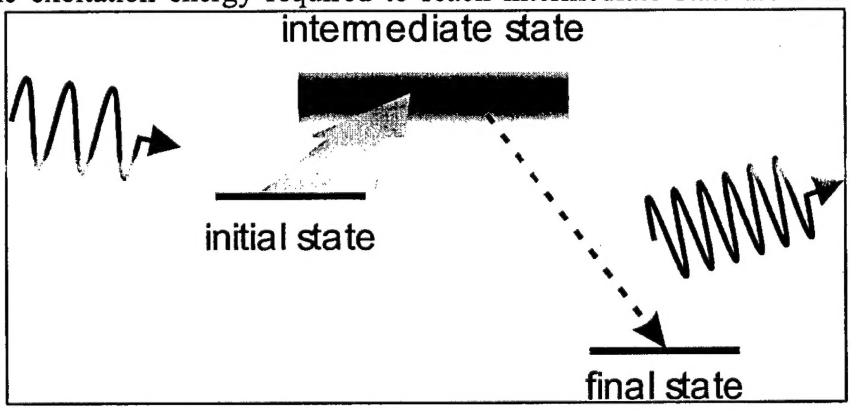

Figure 1. This schematic diagram shows the general process of x-ray driven (triggered) gamma emission if the initial state is an isomer and final state is the ground state. Equivalently, the diagram also represents photoexcitation if the direction of the arrows and the roles of initial and final states are reversed.

There is an ongoing search for isomers other than ^{180}Ta that can also be triggered but have more favorable characteristics in terms of applications. Recent experiments in the United States have focused upon the long-lived isomer $^{178}Hf^{m2}$, which stores 2.445 MeV releasable as gamma rays. A partial energy level scheme for this system is presented in **Figure 2.** More experiments are needed with this isomer, and understanding of the energy release processes must be further developed.

The Principal Investigator, Mr. Hill Roberts, began defining nuclear isomer energy extraction as early as 1991. Since that time SRS Technologies has been directly involved in demonstrating that nuclear isomers can be triggered into an accelerated decay mode to release gamma rays. Mr. Roberts presented a paper in 1995 at GARALAS'95 ("The Importance of Stimulated Gamma Emission from Isomers", *Hyperfine Interactions*, **107** (1997), 91-97) describing the potential of triggered nuclear isomers for many applications. He was a key member of an international team that produced the first triggering data in 1998 (reported in Physical Review Letters in 1999, 82:4 p695). SRS is currently the holder of largest known experimental source in the world of the nuclear isomer $^{178}Hf^{m2}$.

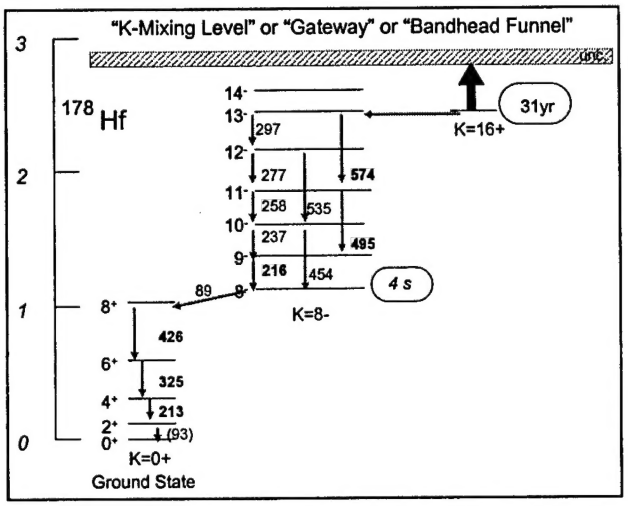

Figure 2. State diagram for ^{178}Hf showing the K=16+ m2 isomer level, K mixing level, and normal decay transitions to ground state.

SRS, with Dr. Carroll of YSU, arranged the use of the X15A monochromatic synchrotron X-ray beam at the BNL NSLS. The goal was to search for the precise triggering energy in the $^{178}Hf^{m2}$ isomer. A polycarbonate encapsulated $^{178}Hf^{m2}$ source was constructed using isomer material previously obtained by SRS. A methodology for the precision survey was developed and implemented in a series of experiments conducted on September 4 – 9, 2001 in collaboration with Youngstown State University. The results were reported at the Quantum Nucleonics session of Lasers01 in Tucson, Arizona in December 2001. The report, "Gamma Spectroscopy of $^{178}Hf^{m2}$ Using Synchrotron X-Rays," by Hill Roberts, Mike Helba, Jeff Carroll, J. Burnett, T. Drummond, J. Lepak, Zhong Zhong, and Jack Agee was published as proceedings in *Hyperfine Interactions* in 2002.

These experiments probed the hafnium sample with incident photon energies over the L_1, L_2, and L_3 X-ray edges of hafnium and the 12-13 keV range. Resonances larger than the experimental minimum detectable level of 10^{-25} cm^2 keV were not observed.

Additional experiments were conducted by SRS and YSU at the NSLS in February and March 2002. These experiments used new encapsulated $^{178}Hf^{m2}$ targets developed by SRS. These targets sandwiched the hafnium isomer between two beryllium windows in an aluminum frame. One of the targets included a water cooling system to control the temperature during high photon flux experiments.

Three experiment types were conducted. The first experiment was basically a duplication of the 1998 experiment with much more favorable conditions. The X15A synchrotron beam was used in the white beam mode. Spectra were collected with the beam on and the beam off. The target was mounted on a precision motorized stage. This allowed the target to be moved minutely to collect a third spectrum in which the synchrotron beam passed through the beryllium window of the target but did not strike the hafnium deposit.

The second experiment used the Youngstown State University Miniball gamma calorimeter to perform time-energy resolved calorimetry and spectroscopy during irradiation in the X15A in monochromatic mode. The Miniball includes six NaI scintillation detectors and one HPGe detector to collect data in a near-4π configuration.

The third experiment was designed to demonstrate a significant degree of isomer "burn up" in the target. The goal was for the accelerated decay to be of such a magnitude that the difference in source activity measured before and after irradiation should be substantially greater than the difference accountable to the ordinary 31-year decay rate for the isomer. For this experiment we used the X17B1 wiggler beamline at the NSLS with the water-cooled target. The target was kept in the beam for four days. A precise assay was conducted in the SRS low-background spectroscopy laboratory before the target was sent to NSLS. Post-irradiation assays of the burn-up target and a control target were conducted in the months following the irradiation.

2.0 Objectives

The objectives for this effort as stated in our proposal are repeated below:

1. SRS will complete the analysis and presentation of data obtained from three experiments conducted at the Brookhaven National Laboratory National Synchrotron Light Source in February and March 2002. These three experiments are referred to as White Beam Spectroscopy, Monochromatic Miniball Cascade Search, and Burn-up Demonstration. We will complete the analysis of these experiments in cooperation with Dr. James J. Carroll of Youngstown State University, who is separately funded.
2. SRS will develop an improved experimental protocol for a definitive burn-up experiment. We will determine the optimal gamma spectrometer settings and assay procedures based on trade studies and exploration of tool capabilities. We will begin collection of long-term pre-burn-up assay data to characterize the burn-up and control targets with the new spectrometer settings.
3. SRS will support other isomer researchers as directed by the Government within the constraints of funds available under this contract. As part of this support, SRS can supply researchers with $^{178}Hf^{m2}$ targets from its limited supply of targets which are currently stored in the SRS facility or already loaned to other researchers, subject to availability and GFE approval from AFRL.

3.0 Results

By the time this research was authorized to proceed, the analysis of the white beam spectroscopy and the monochromatic miniball cascade search had already been completed by YSU under separate funding vehicles. These results have been reported separately. To summarize, neither of these studies were able to identify any statistically significant evidence of isomer triggering during these experiments. However, it should be noted that the miniball apparatus was not fully functional in terms of its electronic support systems and hence the data has proven to be inadequate for any further analysis. Similarly, the white beam experiments were performed by collecting only 20 seconds of data at each energy. An example of this data is presented in **Figure 3**. At the time of these

experiments it was erroneously expected, based upon reports from the UTD group, that relatively large percentage increases (>10%) might potentially be observed during triggering events. These experiments were designed to detect such events. However, since that time it has become apparent that much longer irradiation times would be required to reduce the measurement uncertainties to levels sufficient to detect triggering enhancements of only a few percent. Therefore, no further search for triggering events in the data was performed. Triggering at levels of less than 5% would be hopelessly buried in the measurement noise.

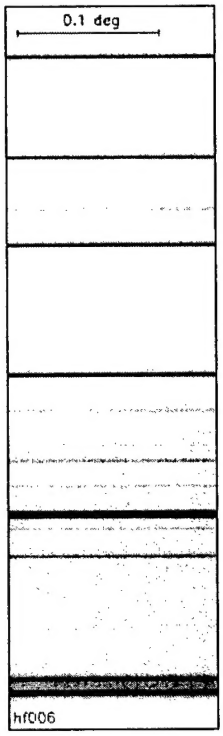

Figure 3. This is an example of multiple spectra collected at NSLS. The vertical axis shows increasing spectrum energies from 46 keV to 613 keV. The horizontal axis shows decreasing incident photon energies from 12.6 keV to 12.4 keV. Gamma lines that are always present appear as constant horizontal lines. A new peak that only appears at one particular incident energy would appear as a dark dot on the graph.

This leaves the burn-up experiment for further analysis in this effort. At the time of the March burn-up experiment, there were no funds available for conducting data analysis. The experiment itself was privately funded in its entirety with SRS funds. With this BAA we have now been able to perform this analysis, which is reported below.

3.1 Burn-up Demonstration

Background: The Burn-up Demonstration was designed to demonstrate a significant degree of isomer "burn up" in the target. For this experiment we used the X17B1 wiggler beamline at NSLS with the water-cooled target. The setup is shown in **Figure 4**. The target was kept in the beam for 97 hours. A precise assay was conducted in the SRS low-background spectroscopy laboratory before the target was sent to NSLS. Post-irradiation assays of the target and a control target were conducted in the months following the irradiation. If the accelerated decay caused by the irradiation is of sufficient magnitude, the difference in source activity measured before and after irradiation should be substantially greater than the ordinary 31-year decay rate for the isomer.

Analysis of the data from the assays revealed that a detrimental shift in the calibration of the gamma spectrometer in the SRS Gamma Spectroscopy Laboratory occurred during the course of this experiment. However, certain steps, such as the use of a control target,

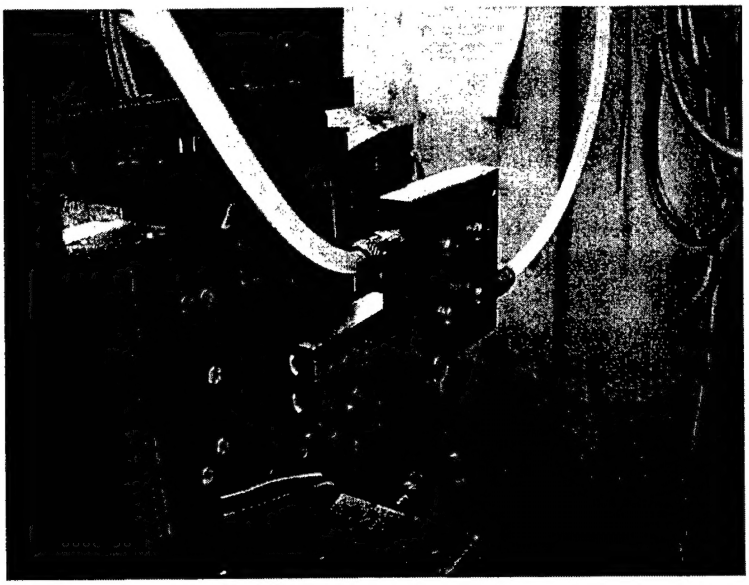

Figure 4. An SRS isomer target for high intensity synchrotron irradiation is shown here in the X17B1 target hutch at the NSLS. This target consists of 1.26×10^{13} $^{178}Hf^{m2}$ isomers deposited as a chloride salt in a 3 mm spot onto a 1 x 25 mm beryllium disk (at center of target). This is covered by a similar beryllium disk. The "sandwich" is clamped together by the outer aluminum housing, which is water cooled.

have allowed the extraction of useful information from the data. The preliminary analysis did not indicate any statistically significant evidence of burn-up.

Approach: SRS Technologies completed the analysis of the Burn-up Demonstration data. One area explored is the question of the half-life of $^{178}Hf^{m2}$. The accepted value is 31 years. However, the community concurs that this value may not be completely accurate. If the half-life is not 31 years, then one of the analysis methods that failed to identify burn-up evidence becomes inaccurate. Additionally, there is a possibility that the half-life may be altered during synchrotron irradiation by interactions with the substrate if the processes are driven by interactions between nuclear and inner shell atomic/electronic states. This would only occur during irradiation. After irradiation the decay rate should be the usual 31 year half-life.

Figure 5 shows the data from this experiment. At first this appears to show a distinct shift in the post-burn-up decay. However, when compared to the control target, which was not irradiated, it is apparent that it also exhibits the same shift in post-burn-up data, **Figure 6**. (The control target was not irradiated, but we still refer to it as "post-burn-up" simply for convenience sake.)

We immediately realized that there was some change in the spectrometer performance which occurred during the time period of the burn-up. During this time the spectrometer (in Huntsville) was shut-down since there were no staff present to maintain it while we were at the Light Source in New York performing the burn-up. Nonetheless, since the shift appeared to affect both the experimental and control targets similarly, we believed it might be possible to determine if a true shift had occurred by normalizing all data to the control target.

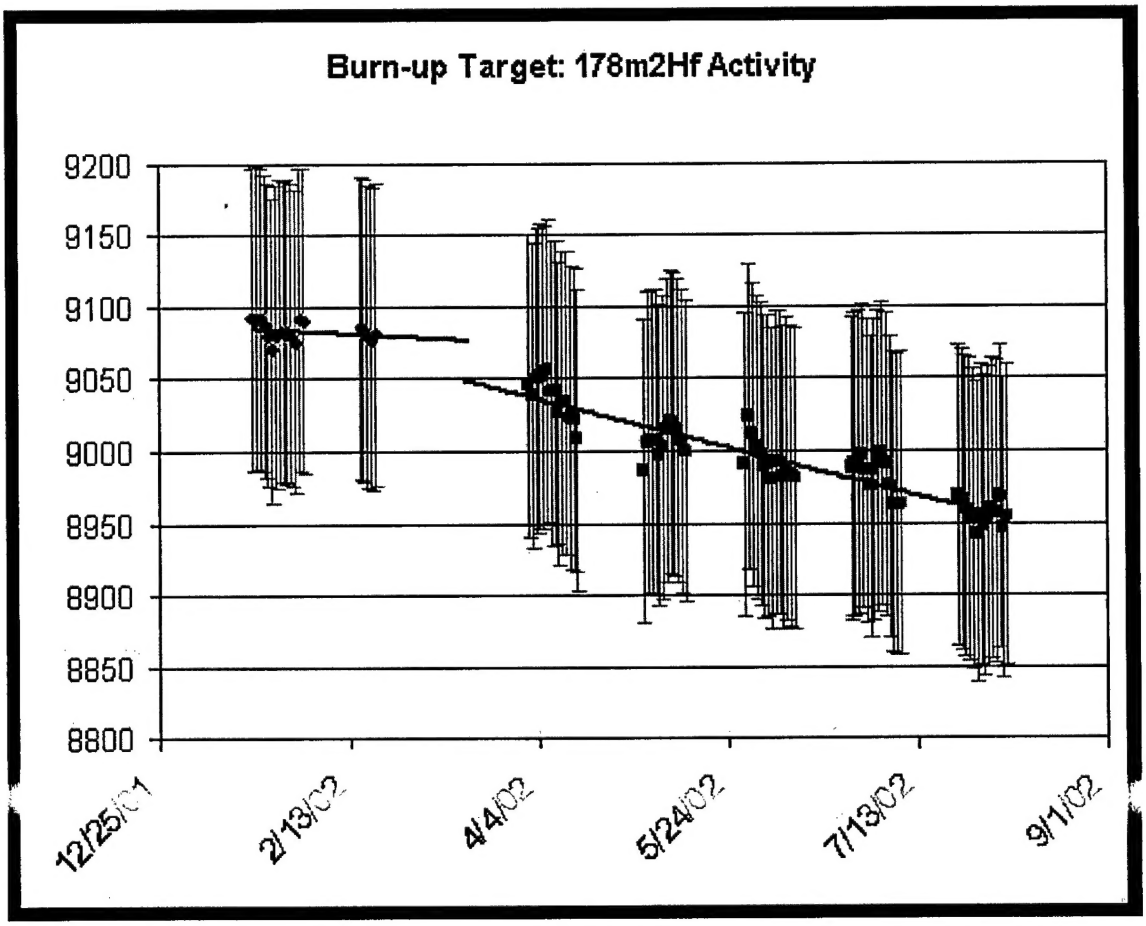

Figure 5. Burn-up Target Hf-178m2 Decay Curve. This shows calibrated decay events as determined using a 10% HPGe detector in a low background measurement chamber. The spectrum was analyzed using the GammaVision software package. Blue indicates pre-burn data. Red indicates post-burn data. The error bars are one sigma. The blank data between measurement groups are when the control target was being measured.

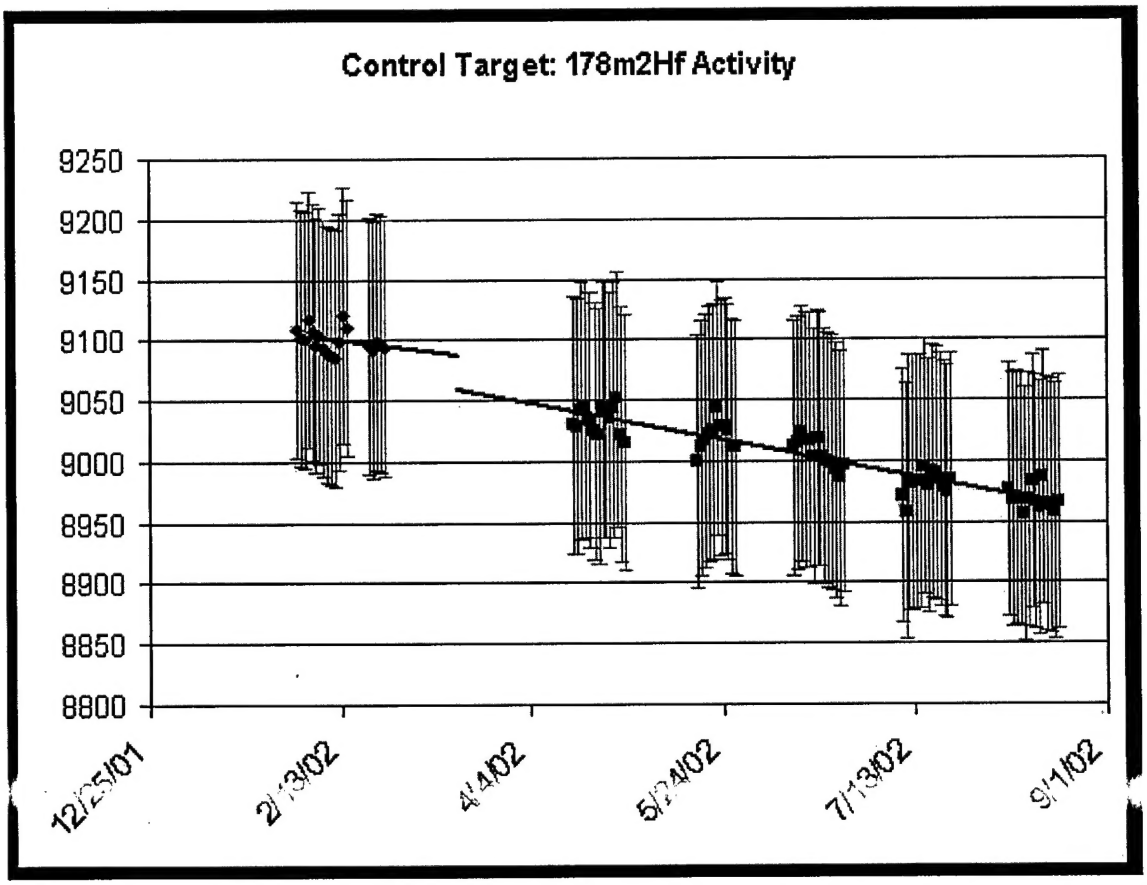

Figure 6. Control Target Hf-178m2 Decay Curve. This shows calibrated decay events as determined using a 10% HPGe detector in a low background measurement chamber. The spectrum was analyzed using the GammaVision software package. Blue indicates pre-burn data. Red indicates post-burn data. The error bars are one sigma. The blank data between measurement groups are when the burn-up target was being measured.

During this analysis effort we evaluated several methods of performing this normalization. The most reliable method was to compare the Hf-178m2 decay to the Hf-172 decay since the Hf-172 decays with a 1.6 year half-life compared to the much slower Hf-178m2 (31 years). **Figure 7** shows the Hf-172 data acquired in parallel with the burn-up data. The Hf-172 is a secondary isotope in these targets which was never separated from the Hf-178. Note that the mid-experiment spectrometer shift is relatively inconsequential with respect to much greater effect of the rapid decay time.

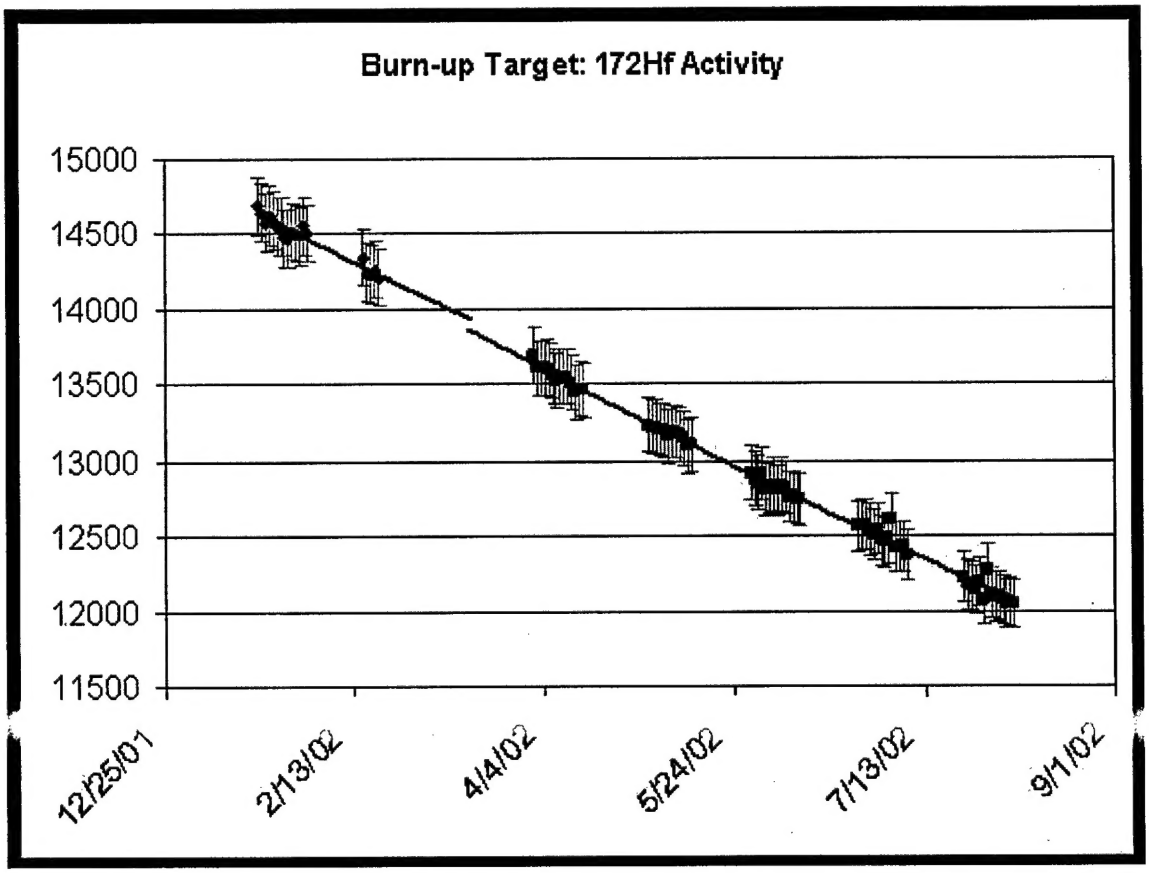

Figure 7. Decay Curve for the Hf-172 component of the Burn-up target. This isotope decays with a 1.87 year half-life. Note that the rate of decay is such that the change in activity over the course of the experiment was substantially greater than the measurement uncertainties. As can be seen by comparison to the Hf-178m2 decay curves, this was not the case for the Hf-178m2.

The results of this normalization method are shown in **Figure 8**. This shows all the data points taken pre and post burn-up. (The burn-up occurs for data points above number 22 in this graphic.) The average of the Hf-178m2 burn-up-to-control ratio is 99.81% for the pre-burn-up and 99.91% for post-burn-up. Similarly, the Hf-172 burn-up-to-control pre and post ratios are 100.08% and 100.25%, respectively. Therefore, we conclude there

was no detectable burn-up of the Hf-178m2 isomer due to the exposure to the synchrotron white beam. This does not mean there was no burn-up, only that this experiment was unable to detect it.

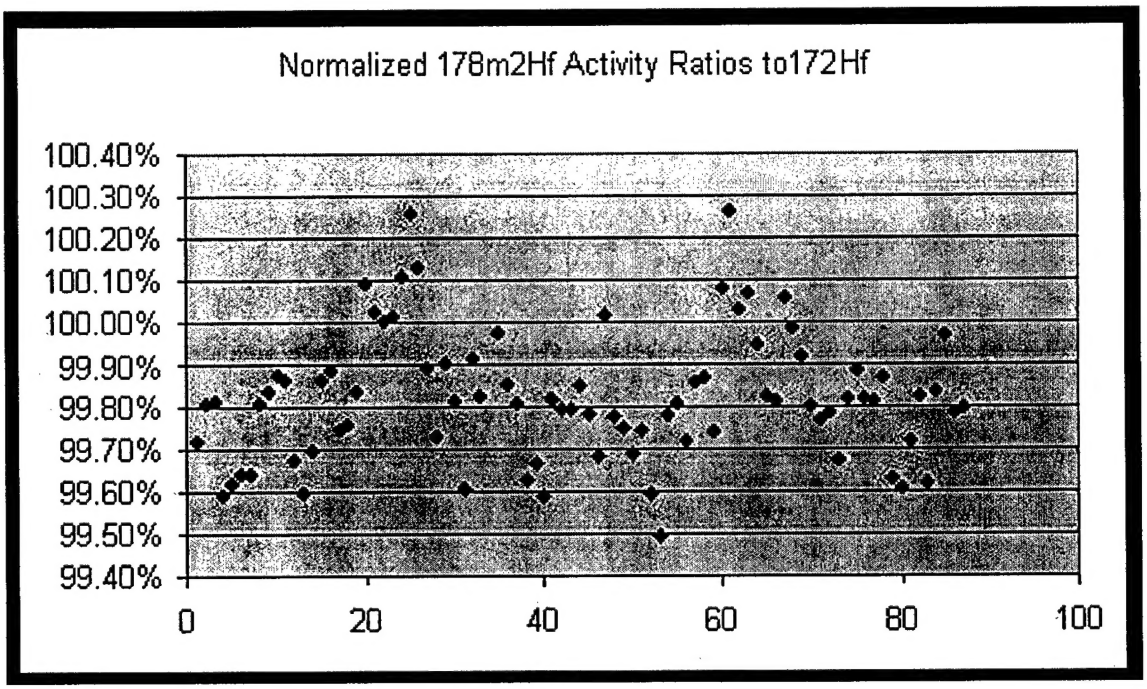

Figure 8. Hf-178m2 activity (normalized to the control target) compared to the Hf-172, by data acquisition sequence number. Notice that all of the data shows essentially no significant variance from 100%, indicating no detectable burn-up is apparent in these data.

The data helps us to identify three major problems with the experiment as conducted. (1) There was insufficient data collected prior to the burn-up and some of the last pre-burn data collected was clearly corrupted by instrumental shifts. This left very little baseline data against which to perform a comparison. (2) The spectrometer should have been operated continuously during the burn-up period and continued to collect decay data from the control target. This might have allowed better calibration and provided a means to correct the erroneous pre-burn data. (3) The irradiation was inadequate to cause a statistically significant burn-up in such a long-lived isomer. This would require a longer in-beam time, or a substantially more intense beam. Neither was available.

Notice that for both the control and burn-up targets the decay curves (Figures 6 & 7) not only show a shift, but also a change in slope. Triggering or burn-up would be evidenced by a step shift in the decay curve, but the slopes would remain the same. The change in slope is a-physical. Such would indicate that the decay half-life changed permanently after the irradiation. However, this apparent slope change is due to erroneous and limited pre-burn data corrupting the pre-burn slope. If the last group of pre-burn data points are eliminated, it is obvious that the remaining pre-burn data would match with the post-burn decay data within measurement uncertainty. These data points are taken to be simply bad data, probably due to the shift with occurred with the spectrometer. This holds for both

the burn-up and control targets. It also occurs with the 172 data, but since this isotope decays so much more rapidly, this becomes a relatively minor error for the 172 data.

These insights provide us direction for establishing a better protocol for a burn-up experiment that might be successful. As yet such an experiment has not been performed and is out-of-scope for this effort.

3.2 Burn-up Experiment Protocol Improvements

The preliminary analysis of the data from the first Burn-up Demonstration revealed several issues that were detrimental to the experiment. Consider **Figure 9**. This shows a putative six month burn of Hf-178m2 undergoing a 10% enhancement of decay during the burn. (Typical results report enhancements in synchrotron beams of 2% or less.) Even with a five-fold increase in enhancement above reported results, it is apparent that a six-month burn will not be sufficient to detect triggering by this method for the Hf-178m2 isomer. For comparison we show in **Figure 10** a similar experiment using the Lu-177m isomer, which has a half-life of 167 days. This becomes a much more feasible experiment. In addition, this isomer can be monitored for beta decay during irradiation which makes a more attractive experimental approach for burn-up analysis.

The primary instrumental issue was a shift in the detector calibration during the time it was not in use while the target was at the NSLS. The repeat experiment will be conducted with a much more rigorous measurement protocols to prevent the deficiencies that occurred in the original experiment.

1. Assays of the burn-up and control targets will be alternated with assays of simpler calibration sources to verify the spectrometer calibration performance.
2. Pre-burn data will be collected over a much longer period comparable with the post-burn up data.
3. The detector will never be turned off. Assays of the control target and calibration sources will continue while the burn-up target is sent to the NSLS.
4. Liquid nitrogen fills will be scheduled for times when no data collection is in progress.
5. Alternative isomers such as Lu-177m will be evaluated with shorter half-lives which are more compatible with available irradiation times.

SRS has identified modifications to the spectrometer and analysis software settings that greatly enhance the quality of the data collected. These modifications fall into two categories. The first category is comprised of modifications intended to improve the accuracy of the activity estimates. For example, true coincidence summing can cause inaccuracies in the spectra obtained by the detector. A true coincidence summing correction to the peak counts can be applied that follows through to the activity estimates. The other category is comprised of modifications intended to reduce the variance of repeated detector measurements. Two examples are the gain and zero stabilization features of the digital spectrometer.

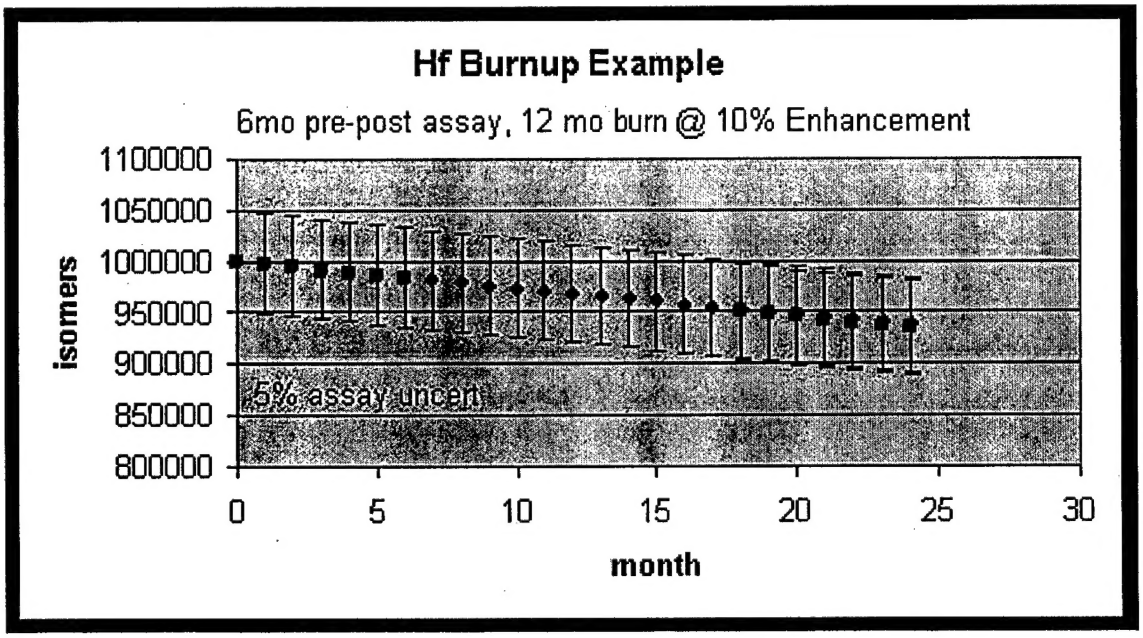

Figure 9. Hypothetical Hf-178m2 Burn-up Experiment during which all decays are enhanced by 10% during a twelve month burn irradiation. Six month pre and post burn assays are assumed with projected measurement uncertainties. Even with these experimental parameters, burn-up is not detectable. This is due to the very slow decay rate, leading to large measurement uncertainties in decay activity changes when measurement times are small compared to the half-life.

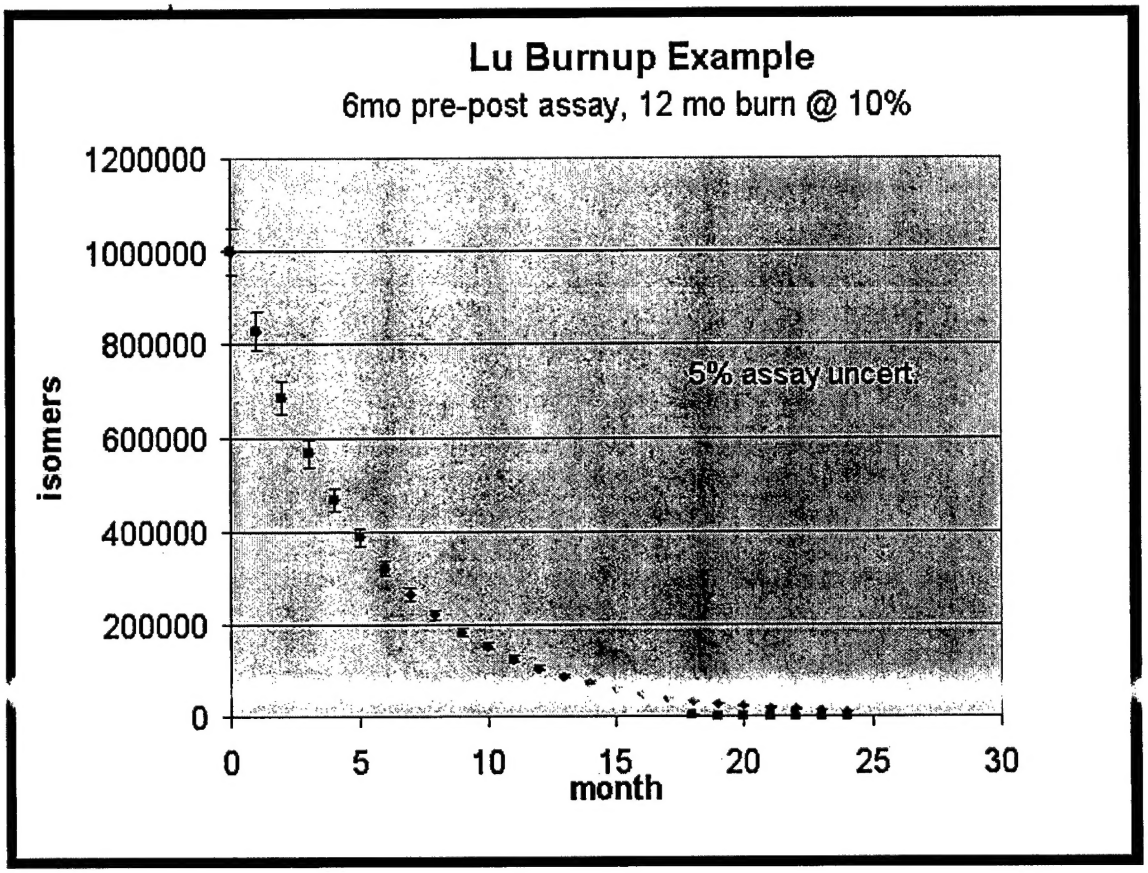

Figure 10. Hypothetical Lu-177m Burn-up Experiment during which all decays are enhanced by 10% during a twelve month burn irradiation. Six month pre and post burn assays are assumed with projected measurement uncertainties. Since the decay half-life (167 days) is comparable to the experiment timeline, burn-up would be expected to be observed for this hypothetical conditions. (To date there is no data indicating Lu-177 is triggered by synchrotron radiation.)

SRS has continued to identify potential spectrometer setting improvements and test them through trade studies. Selected improvements have been documented and implemented in our spectra analysis protocols.

SRS has established new data analysis techniques, and some of these have been identified during the Burn-up Protocol task to streamline their implementation. For example, certain gamma peaks are more beneficial to analyze than others. By identifying these peaks before the assay begins, the spectrometer can be programmed to automatically output reports documenting the lines of interest after each assay interval.

3.3 Isomer Experimental Support

SRS provided three weeks of experimental support to an experiment conducted by Dr. Carroll of YSU at Spring8 during June of 2004. SRS also provided the $^{178}Hf^{m2}$ target for this experiment, as well as our on-site support. Mr. Mike Helba provided the on-site support. (Mr. Roberts was at another venue to present these results at Dr. Agee's isomer workshop section of the Frontiers of Non-linear Physics conference.) **Exhibit 11** shows some of the targets created by SRS. At the time of this report the data are still being analyzed by Dr. Carroll and will be reported separately to AFOSR under the primary contract vehicle supporting Dr. Carroll from AFOSR. Preliminary analysis shows some indication of potential triggering events, but at a level that is inclusive pending rigorous data analysis.

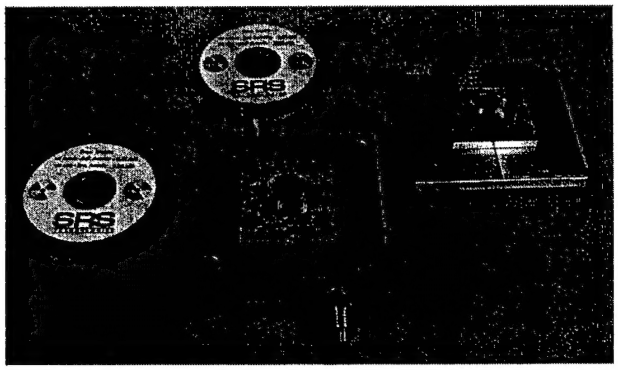

Exhibit 11. These are examples of $^{178}Hf^{m2}$ experimental sources developed by SRS. The polycarbonate and Lucite targets are on the left and top. The beryllium "sandwich" sources are on the right and bottom. The bottom source is water cooled for high photon intensity applications.

4.0 Conclusion

The data from the March 2001 Burn-up experiment has now been fully analyzed. No burn-up due to xynchrotron radiation was detected in this experiment due to both experimental limitations and the unlikelihood of ever observing burn-up in the Hf-178m2 system for triggering enhancements of less than 10%. We suggest looking at alternative isomer systems with shorter half-lives, such as the Lu-177m isomer.